ndian J. Phys. 75A (2), 109-111 (2001)

Rapid Communication

IJP A
— an international journal

Observation of successive phase transitions in ferroelectric liquid crystal (CS-1013) from dielectric measurements and a pseudospin type model for studying ferro and antiferroelectric behaviour

S K Kundu, E Okabe[†], B K Chaudhuri* and W Haase**

Solid State Physics Department, Indian Association for the Cultivation of Science
Jadavpur, Calcutta-700 032, India
[†]Specialty Chemicals Research Center, Chisso Petrochemical Corporation, 5-1
Goikaigan Ichihara Chiba 290-8551, Japan
**Department of Physical Chemistry, Technical University Darmstadt,
Petersen strasse-20, Darmstadt, Germany

E-mail sspbkc@mahendra.iacs.res.in

Received 13 November 2000, accepted 23 November 2000

Abstract : From dielectric measurements, we have shown the appearance of successive phase transitions in a ferroelectric liquid crystal CS-1013 between crystalline and isotropic phases, we observed successive phase transitions *viz* Cr - ~ 35° SmC* - 63°C - SmA 70°C-N* 80°C-Iso Both Goldstone mode (GM) and Soft mode (SM) are observed in the SmC* phase A theoretical pseudospin type model has been suggested for studying ferro and antiferroelectric phase transitions in liquid crystals

Keywords Liquid crystal, ferroelectricity, Goldstone and soft mode

PACS No. 77.90.+k

Phase transition in crystalline ferroelectric material is a vastly studied field of interest [1]. But phase transition in ferroelectric liquid crystal is a most recent discovery when 4-(1-methylhepty loxycarbonyl) phenyl 4'-octylbiphenyl-4-carboxylate (MHPOBC) was found to be the first ferroelectric liquid crystal [2] After this discovery, many chiral compounds with mesophases *viz.* ferroelectric (FE)-SmC*, antiferroelectric (AFE)-SmC_A*, ferrielectric (FEE)-SmC_γ, $Sm_\alpha C$* etc. have been discovered by electro optic [3, 4], switching [2, 5, 6], pyroelectric [7] and other measuring techniques. However, none of these techniques is self-sufficient to provide detailed information of the ferroelectric phase transition, molecular dynamics *etc.* Temperature and frequency-dependent dielectric permittivity and loss data provide important information about the successive phase transitions and molecular dynamics in liquid crystals.

In the present communication, we report the different phase transitions occurring between the crystalline and the isotropic phases of a typical ferroelectric liquid crystal FLC *viz.* CS-1013 using dielectric spectroscopic technique. The sample was prepared by Chisso Petrochemical Corporation, Japan. Dielectric constants (both real and the imaginary parts) were measured by a computer controlled HP 4192A impedance analyzer having measuring frequency range between 5Hz and 13MHz. The temperature was controlled (with an accuracy of ± 0.1°C) by an Eurotherm temperature controller The temperature range of our investigation was between 29-82°C. Our experimental set-up is similar to that described elsewhere [8, 9]. Sputtered gold plated electrodes were used to measure the dielectric constant of the FLC layer of thickness ~ 15 μm. The FLC filled cell was prepared by capillary action method in an isotropic phase. Stabilized low frequency ac field (~ 1.67 kV/cm) was used for better molecular alignment in this FLC sample. The different phases of SC-1013 studied by other methods [10] are shown below

	U.K.	63°C	70°C	80°C		
Crystalline	---------	SmC*	-------	SmA	---------	N*	--------	Iso	(1)

Some important parameters of the liquid crystal under investigation are shown in Table 1.

* Corresponding Author.

Table 1. Some important parameters of the ferroelectric liquid crystal CS-1013

Spontaneous polarization (Ps)	25°C	15 0nCcm^{-2}
Tilt angle (θ)	25°C	26 deg
Helical pitch in N* phase	- T_N[1]	38 μm
Helical pitch in SmC* phase	25°C	6 μm
Response time (τ)	25°C, E = ± 10V/μm	739 μsec
Rotational Viscosity (η_0[2])	25°C	1282 mPa s
Optical anisotropy (Δn)	25°C, λ = 546 nm	0 12

1) T_N means N*-SmA just above transition temperature

2) $\eta_0 = \eta / \sin^2\theta$

Figure 1 shows the temperature dependent dielectric loss (ε'') factor at two different fixed frequencies (1 and 10 kHz). Different phase transitions described by eq. (1) are clearly observed from this Figure. The SmA-SmC* phase transition appears to be first order in nature. Temperature dependence of dielectric permittivity (ε') shown in Figure 2 also indicates the presence of different phases in SC-1013.

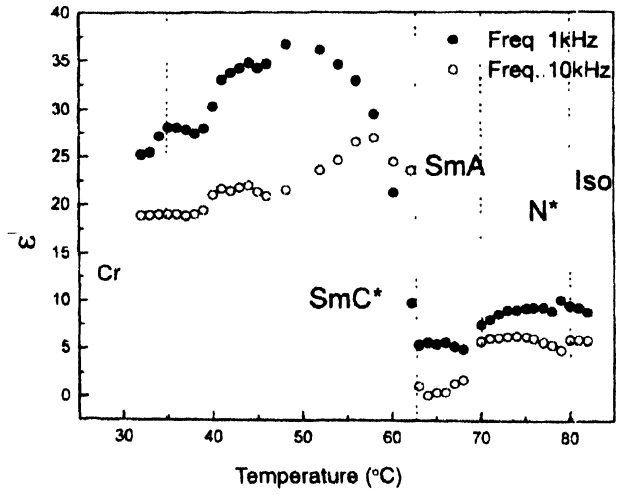

Figure 1. Temperature-dependent dielectric loss (ε'') of ferroelectric CS10-13 showing different phases

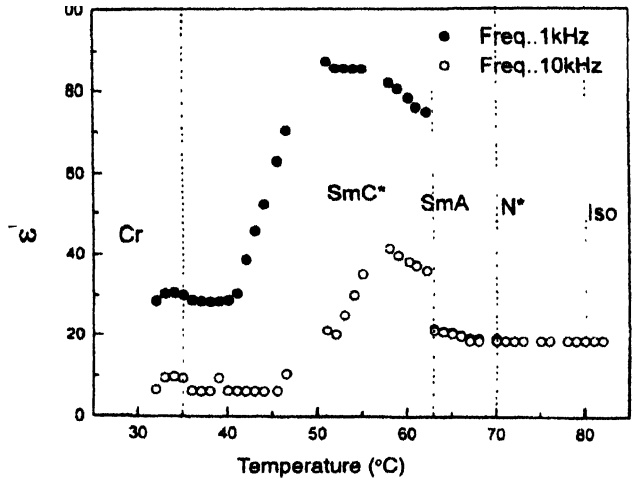

Figure 2. Temperature-dependent dielectric permittivity (real part ε') of ferroelectric CS-1013 at two different temperatures.

It is well known that in planar alignment, the dielectric permittivity due to Goldstone mode [8, 11] is dominated in SmC* phase which appears due to the phase fluctuations in tilt angle of the molecules. The other electric permittivity contributions are due to soft mode [12] (caused by amplitude fluctuation in tilt angle), domain mode [13] and unwinding mode [14] (earlier theoretically predicted). These modes are generally observed by suppressing the Goldstone mode in SmC* phase. In the present sample, the dielectric permittivity due to Goldstone mode is dominated in the SmC* phase. However, soft mode is also observed near the SmA-SmC* transition region, as observed from Figure 3. It is interesting to note that one also observes (from Figure 3) a new molecular mode around 70kHz. This new mode is considered to be due to the re-orientation of the long helical axis. Similar curves have also been obtained at different temperatures and frequencies showing several interesting features in this FLC which would be published elsewhere

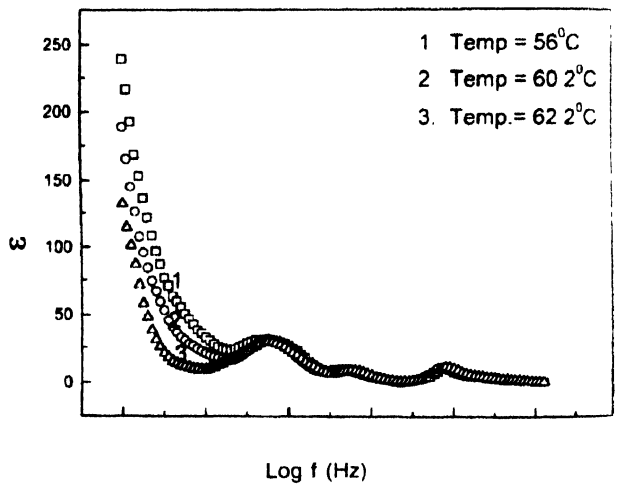

Figure 3. Frequency dependence of the imaginary part of dielectric constant (ε'') in ferroelectric SmC* phase at different fixed temperatures

We find that the crystalline to ferroelectric (SmC*) phase transition can be well described by a pseudospin Hamiltonian of the form

$$H = -\mu E \Sigma \sigma_i^z - U\Sigma(\sigma_i^z)^2 - B\Sigma(\sigma_i^x)^2 - \Sigma J_{ij}\sigma_i^z\sigma_j^z.$$

Here, the pseudospin variable σ (with σ_x, σ_y, and σ components) is associated with the long axis of the molecule μ is the electric field (E)-dependent dipole moment, U is the magnitude of the uniaxiality parameter representing single molecular anisotropy in the z direction of polarization (a measure of the magnitude of pitch), B is the magnitude of the biaxiality parameter representing the molecular anisotropy transverse to the z-direction (a measure of the tilt angle θ) and J_{ij} (>0 for ferroelectric transition and < 0 for antiferroelectric transition) is

the effective exchange interaction between the dipoles (pseudospin). J is also a measure of the interaction strength between B and U (*i.e.* pitch and θ) at sites i and j. All the parameters U, B, and J undergo thermodynamic fluctuations (along with tilt angle and pitch) and are responsible for different mesophase transitions in the liquid crystals. Though this model Hamiltonian [eq. (2)] is similar to that of Devil's staircase model used by Yamashita and Miyazima [15], our derivation of the model and physical significance of the model parameters are quite different from those used by them. Detailed calculations of the transition temperature, spontaneous polarization *etc.* using this model [eq. (2)] will be published elsewhere.

Acknowledgment

One of the authors (BKC) is grateful to the AvH Foundation for providing the HP frequency analyser and the temperature controller as gift. The author SKK is also grateful to the Council of Scientific & Industrial Research (India) for providing a fellowship to work in this project.

References

[1] M E Lines and A M Glass *Principles and Applications of Ferroelectrics and Antiferroelectrics* (Oxford . Clarendon) (1977)

[2] A D L Chandani, T Hagiwara, Y Suzuki, Y Ouchi, H Takezoe and A Fukuda *Jpn J Appl Phys* **27** L729 (1988)

[3] Yu P Panarin, O Kalinovaskaya and J K Vij *Appl Phys. Letts* **72** 1667 (1998)

[4] K Hiraoka, T Tsunita, Y Sugiyama, K Monzen, Y Uematau and Y Suzuki *Jpn J Appl Phys* **36** 6847 (1997)

[5] Y Lee, A D L Chandani, K Itoh, Y Ouchi, H Takezoe, A Fukuda *Jpn J Appl Phys* **29** 1122 (1990)

[6] H Orihara, T Fujikawa, Y Ishibashi, Y Yamada, N Yamamoto, K Mori, K Nakamura, Y Suzuki, T Nagiwara, I Kawamura *Jpn J Appl Phys* **29** L333 (1990)

[7] J W O'Sullivan, Yu P Panarin, J K Vij, A J Seed, M Hird and J W Goodby *Mol Cryst Liq Cryst* **301** 189 (1997)

[8] A M Birader, S Wróbel and W Haase *Phys Rev* **A39** 2693 (1989)

[9] W Haase, H Pranoto and F -I Bormuth *Ber Bunsenges Phys Chem* **89** 1229 (1985)

[10] T Minato and K Suzuki *Liq Cryst* **23** 143 (1997)

[11] M Pfeiffer, G Soto, S Worbel, W Haase, R Twieg and Betterton *Ferroelectrics* **121** 55 (1991)

[12] S Worbel, A M Biradar and W Haase *Ferroelectrics* **100** 271 (1989)

[13] W Haase, S Hiller, M Pfeiffer and L A Beresnev *Ferroelectrics* **140** 37 (1993)

[14] B Zeks, T Carlsson, I Musevi and B K Urbane *Liq Cryst* **15** 103 (1993)

[15] M Yamashita and S Miyazima *Ferroelectrics* **148** 1 (1993)